



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of : **Confirmation No. 6230**  
Daisuke ITOH et al. : Attorney Docket No. 2005\_0470A  
Serial No. 10/528,229 : Group Art Unit 1794  
Filed March 18, 2005 : Examiner Ellen S. Wood  
**STRONGLY STRETCHED ALIPHATIC : Mail Stop: AMENDMENT**  
**POLYESTER MOLDINGS**

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**SUPPLEMENT TO INFORMATION DISCLOSURE STATEMENT**

Commissioner for Patents  
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Sir:

Referring to the IDS filed December 22, 2008, Applicants will now discuss References (1)-(8) identified at the top of page 3 of the IDS.

Please note that Ref. (1) is a Japanese counterpart of the Mitsuhiro et al. reference (EP '221) applied by the Examiner to reject the claims of the present application in the Office Action of October 16, 2008.

Ref. (3) includes Examples 1- 3, wherein films of a mixture comprising an aliphatic polyester (A) selected from polyhydroxybutylate, polyhydroxyvalerate and a copolymer of hydroxybutylate and hydroxyvalerate, and an aliphatic polyester (B) selected from polycaprolactone, and homo- or co-polymers of glycolic acid, lactic acid, etc., are subjected to longitudinal stretching at ratios of 3.8 - 4.0 times at 55 °C, and then to lateral stretching at ratios of 3.5 - 4.0 times at 55 °C.

Similarly to Ref. (3), Ref. (4) includes Examples 4 – 6, wherein films of a mixture comprising a copolymer of 3-hydroxybutylate and 3-hydroxyvalerate, and poly ( $\epsilon$  - caprolactone), are subjected to longitudinal stretching at a ratio of 4.0 times at 55 °C, and then to lateral stretching at a ratio of 3.6 times at 55 °C.

These references both fail to disclose Tg of the polymers and fail to disclose or suggest any intense stretching effect. Moreover, the aliphatic polyesters used in these references are clearly different from the specific glycolic acid polymer recited in amended claim 1 of the present application.

Ref. (5) includes Examples 1 and 2, wherein glycolic acid homopolymer (Example 1) or glycolide/lactide (98/2 by weight) copolymer (Example 2) is subjected to blow molding including successive biaxial stretching longitudinally at 2.23 times, and then to laterally (diametrically) stretching at 2.81 times. These stretching conditions are milder than biaxial stretching of vertical x lateral stretch ratios of 3 x 3 times in Comparative Example 1 of the instant specification and, similarly to Comparative Example 1, intense stretching effects intended by the present invention cannot be expected.

Ref. (6) includes Examples 1– 8, wherein films of polylactic acid containing 5 – 30 wt.% of plasticizer are subjected to longitudinal stretching at 3 times at 50°C and, after application of an acrylic-type aqueous dispersion coating, to lateral stretching at 5.0 times. It is clear that the intense stretching effects of the present invention cannot be expected by the stretching of aliphatic polyester containing such a large amount of plasticizer.

Ref. (7) discloses a method of elevating the melting point of polycaprolactone by subjecting a mixture of polycaprolactone with a thermoplastic resin having a melting point higher than that of polycaprolactone to stretching at a temperature around the melting point (ordinarily 55 – 60°C) of the polycaprolactone. The stretching temperature is much higher than the Tg(= around – 60°C) of polycaprolactone, and the disclosed method of elevating the melting point of polycaprolactone of Ref. (7) is clearly different from the method of elevating

the crystal melting point of aliphatic polyester through intense stretching of the present invention. Moreover, the melting point-elevating effect of Ref. (7) is described to be peculiar to polycaprolactone (paragraph [0008]).

Finally, Ref. (2) and Ref. (8) (prior application, counterpart U.S. publication not found) include Examples wherein a 80mm x 80mm film of a glycolic acid copolymer comprising 78 – 90 mol% of glycolide and 10 – 22 mol% of another aliphatic ester dimer such as lactide is subjected to simultaneous biaxial stretching at a stretching speed of 50%/min. of vertically 3.5 times x laterally 3.5 times in a chamber of 65°C (Example 1 of Ref. (2)) or 60°C (Example 1 of Ref. (8)).

In consideration of the glycolide molecular weight of 116 and lactide molecular weight of 144, the lactide content in the glycolic acid copolymer of Ref.(2) (or Ref. (8)) is much higher than the glycolic acid copolymer recited in the instant claim 1 obtained by copolymerization of at least 90 wt.% of glycolide and at most 10 wt.% of lactide. This relatively higher lactide content in the glycolic acid copolymer of Ref. (2) is never changed even at a slightly higher copolymerization speed of glycolide than lactide as shown in a copolymerization example described hereinbelow.

In order to confirm this, Applicants prepared a glycolide/lactide (=90/10 by weight) copolymer and subjected it to simultaneous biaxial stretching under the conditions of Example 1 of both Ref. (2) and Ref. (8). The procedure and the results are described below.

#### (Preparation of a copolymer)

In view of the larger copolymerization speed of glycolide, a monomer mixture of glycolide/lactide =88/12 (by mol) (=85.5/14.5 (by weight)) together with 300 ppm of catalyst:  $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$  was sealed up within a tubular bulk-polymerization apparatus as used in the Examples of WO 03/006521A1 and heated for 3 hours at 180°C, and then for 4 hours at 200°C to substantially complete the co-polymerization. The thus-obtained copolymer exhibited a molecular weight (based on polymethyl methacrylate as measured by GPC measurement using

hexafluoroisopropanol solvent) of ca. 220,000; a melt-viscosity of 1000 Pa • s at 240 °C and a shear rate of 120 sec<sup>-1</sup>; and a glycolide/lactide copolymerization ratio of substantially 90/10 (by mol) (=87.9/12.1 (by weight)) calculated from the <sup>1</sup>H-NMR spectrum. The copolymer also exhibited a glass-transition temperature (T<sub>g</sub>) of 41°C as measured through a temperature cycle including heating at a rate of 20°C/min. from -10°C, holding at 270°C for 5 min. and cooling at a rate of 20°C/min.

#### (Stretching test)

The above-obtained copolymer in 100 wt. parts together with 0.1 wt. part of phosphite-type anti-oxidant ("PEP-8", made by Asahi Denka Kogyo K.K.) was pelletized in the same manner as in Example 1 of the present application. The pellets were heated for 5 minutes by a heat press machine set at 200°C and shaped by a cold press set at 25°C to form a 350 μm-thick press sheet similarly as in Example 1 of Ref. (2), which was then immediately cut into two 90 mm-square unstretched sheets.

#### <Tests 1 and 2>

The two unstretched sheets were respectively set at a clamp spacing of 80 mm and subjected to simultaneous biaxial stretching vertically ca. 3.5 times and laterally ca. 3.5 times at a stretching speed of 50 %/min. (i.e., 40 mm/min.) similarly to the Examples of Refs. (2) and (8) at temperatures of 65°C (Test 1) and 60°C (Test 2), respectively.

The resultant stretched films were subjected to measurement of crystal melting points T<sub>m</sub> (°C), X-ray orientation degree (%), and main- and sub-dispersion peak temperatures (°C) by viscoelasticity measurement similarly to the Examples of the present specification. The results are shown in Table A below.

#### <Tests 3 and 4>

The above-obtained glycolide/lactide(=90/10(by mol)) copolymer was further pelletized and formed into two cast sheets (each in a thickness of 100 μm), which were respectively set at clamp spacing of 80 mm and subjected to simultaneous biaxial stretching of vertical x lateral

=4.0 x4.0 times (Test 3) and vertical x lateral =4.5 x4.5 times (Test 4) , respectively, at a stretching speed of 7 m/min. (i.e., 7000 mm/min.), and at 60°C similarly to the Examples of the present specification.

The crystal melting points  $T_m$  (°C) and X-ray orientation degree (%) measured with respect to the thus stretched films are shown in the following Table A together with the results of the above Tests 1 and 2 and Comparative Example 6 of the present specification.

[Table A]

	Polymer	Stretch ratio (-)	Stretch speed	$T_m$	X-ray orientation	DMA	
		(Temp.)	(mm/min.)	(°C)	(%)	Main (C)	Sub (C)
Comp. Ex. 6	PGA homopolymer	Non-stretched	-	218	0	45	-49
Comp. Ex. 1	PGA homopolymer	3 x 3 (45°C)	7000	218	82	63	-47
Control of Tests 1 -4	glycolide/lactide =90/10(by mol) copolymer	Non-stretched	-	195	0	46	-72
Test 1	"	3.5 x 3.5 (60°C)	40	196	-	57	-60
Test 2	"	3.5 x 3.5 (65°C)	40	196	78	58	-58
Test 3	"	4.0 x 4.0 (60°C)	7000	198	90	N.M.*	N.M.*
Test 4	"	4.5 x 4.5 (60°C)	7000	198	91	N.M.*	N.M.*

\*: N/M = Not measured

As shown by the results in Table A, Tests 1 and 2 performed with simultaneous biaxial stretching under the conditions taught in Refs. (2) and (8), provided an increase in crystal melting point by only 1°C and failed in providing the intense stretching effect (an increase in crystal melting point of at least 3°C) of the present invention. This is believed to be because the stretching ratios of vertical x lateral =3.5 x 3.5 times (= areal ratio of 12.25 times) were insufficient.

Tests 3 and 4 using the stretching ratios of vertical x lateral =4.0 x 4.0 times (=areal ratio of 16.0 times) or the stretching ratios of vertical x lateral =4.5 x 4.5 times (=areal ratio of 20.25 times) managed to achieve the increase in crystal melting point of at least 3°C, but is substantially smaller than the increases of 8 – 9°C achieved in Examples 1– 4 with respect to

glycolic acid homopolymer. This is presumably because the glycolide/lactide =90/10 (by mol) (=87.9/12.1 (by weight)) copolymer obtained by using a monomer mixture at a ratio of glycolide/lactide =85.5/14.5 (by weight), which is substantially lower than the lower limit of glycolide/lactide =90/10 (by weight) in instant claim 1, had lower crystallinity and stretch orientation characteristics than the glycolic acid homopolymer, and thus failed to exhibit the intense stretching effects obtained by the glycolic acid homopolymer.

Thus, Refs. (1)- (7) all fail to disclose or suggest the intense stretching effects as represented by an increase in crystal melting point with respect to a glycolic acid polymer having a high glycolide unit content, and thus showing a high gas-barrier property. Only Ref. (8) (prior Japanese application) discloses an improved heat resistance which is at first glance similar to the intense stretching effect of the present invention, but is still within a level attributable to an increase in orientation of an amorphous portion, and does not disclose the improvement in crystal property as represented by an increase in crystal melting point. Further, from the viewpoint of polymer composition, Ref. (8) does not disclose intense stretching effects as represented by an increase in crystal melting point with respect to a glycolic acid polymer having a high glycolide unit content and thus showing a high gas-barrier property.

Respectfully submitted,

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